THE ORIGINS AND PROPERTIES OF ENVIRONMENTAL TOBACCO SMOKE

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Environmental tobacco smoke (ETS) is formed from cigarettes as sidestream and exhaled mainstream smoke diffuse into ambient air. Detailed studies are reviewed which describe how sidestream smoke is formed, its acceleration away from the cigarette and its chemical properties. As the smoke streams diffuse into the atmosphere they become greatly diluted and physical and chemical changes occur. A quarter of the material in sidestream particles evaporates, so that ETS nicotine is virtually entirely in the vapour phase, and the particles shrink. As cigarettes are smoked, the levels of ETS components rise and then fall exponentially due to air exchange and deposition of smoke particles onto surfaces. The decay of ETS also depends on the particular component, with nicotine decaying faster than other substances. In real-world environments, ETS is found along with chemicals and particles from many sources. Studies are reviewed which quantify the contribution of ETS to various indoor air environments. These include determination of the ETS proportion of total respirable particles, measurement of nicotine as a specific ETS marker, and comparisons of chemicals present in matched smoking and nonsmoking environments. The ETS contribution of volatile organic compounds in air is much less than that from other sources. The review emphasises the need for tobacco specific analytes to be used as ETS markers and/or to apportion ETS particulate matter from total particulate matter in the atmosphere.

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INTRODUCTION

Environmental Tobacco Smoke, ETS, has received considerable attention in recent years. ETS is the complex mixture of chemicals found in air as a specific result of tobacco smoking (Nystrom and Green 1986). Some reports have claimed that exposure to ETS can be harmful to the health of nonsmokers (USSG 1986; NRC 1986; ISCSH 1988). This issue has been discussed by scientists and epidemiologists for over a decade and although knowledge has increased over this period, it is still the subject of scientific controversy (Mantel 1987; Uberla 1987).

In order to assess ETS properly it is necessary to understand some of its chemical and physical properties, and to ascertain the concentrations of ETS present in typical environments (Repace 1987b; Proctor et al. 1989a).

The aims of this paper are to describe the origins of ETS through an understanding of the combustion processes occurring within a cigarette, how it builds up and then decays in indoor air, and some of its properties. This paper considers ETS in relation to cigarettes, rather than that originating from cigars and pipes.

THE ORIGINS OF ETS

ETS results from a combination of sidestream smoke (that which is released from the lit end of the cigarette), and exhaled mainstream smoke (that exhaled by the smoker after drawing on the cigarette), both greatly diluted by the ambient air.

Fresh sidestream smoke

Sidestream smoke is defined as all the smoke generated by a cigarette that is not mainstream. Side-

stream smoke is made up of the sidestream plume which is emitted from the burning zone during both the puff and smoulder periods in an upwards direction (because of buoyancy), the smoulder stream which escapes from the mouth end of the cigarette during smoulder, and gases which diffuse out of the tobacco rod by diffusion during both the puff and smoulder periods (Lipp 1965; Hoegg 1972; Baker 1982). It has been estimated that the sidestream plume contributes about 95% to the total sidestream smoke (Hoegg 1972).

Physical and chemical aspects of the sidestream plume in the vicinity of the burning zone of a smouldering cigarette are illustrated in Fig. 1. (In both Figs. 1 and 2, % v/v is % volume/volume. It should be noted that 1 % v/v = 10 mL/L). The data in this figure were measured in a variety of experiments previously described (Baker 1982; Robinson 1987). The position of the sidestream smoke plume was obtained by carefully photographing the smouldering cigarette under controlled air flow conditions around the cigarette. The position of the gas phase plume was obtained in a series of experiments in which small sampling probes were placed around the cigarette and connected directly to a mass spectrometer. The temperature distribution was measured using small thermocouples placed around the burning zone. The velocity distribution was measured using a laser Doppler velocimeter technique.

The gas phase temperature distribution outside the cigarette is very similar to that obtained earlier by Neurath et al. (1966). The positions of the smoke and gas phase plumes have also been confirmed using a Schlieren optical method described by McRae and Jenkins (1987).

In the smoulder period between the puffs, a natural convection flow of air around the burning zone in an upwards direction (because of buoyancy) sustains burning but at a much lower intensity than during the puff. Little change occurs to the external temperature and oxygen distributions when the puff is taken (Baker 1982), indicating that the natural convection stream around the burning zone and into the sidestream plume is only slightly affected by the influx of air during the puff. The combustion processes occurring on the surface of the burning zone in the convection stream proceed independently of those inside.

The main products in the gas plume are carbon monoxide, carbon dioxide, hydrogen and water — the concentration distribution of carbon monoxide outside the burning zone during smoulder is shown in Fig. 1 and the forms of the profiles of the other gases and oxygen depletion are similar. The carbon

monoxide plume originates some 3 to 4 mm in front of the paper burn line, as do the external temperature contours. The carbon monoxide concentration immediately above the burning zone is higher than that just inside, and a similar situation exists for carbon dioxide. Thus, sidestream carbon monoxide and carbon dioxide are not formed only by the combustion products diffusing out of the burning zone — some must be formed on the external surface from reactions with the oxygen convected around the coal.

Fig. 2 illustrates the variations during the smoking cycle of the gas concentrations of oxygen, carbon dioxide, carbon monoxide and hydrogen at a specific point in the sidestream gas plume. This point is situated 1 mm above the surface of the burning zone, and 3 mm in front of the paper burn line. The plume hydrogen and carbon monoxide concentrations at this point increase during the puff while the carbon dioxide concentration falls. At the end of the puff, the level of all three products increases for about one second. This variation is very similar to that found inside the centre of the burning zone (Baker 1981), and is due to the outward diffusion of those products formed inside the burning zone. The small rise in carbon monoxide and fall in carbon dioxide during the puff are due, at least partly, to the carbonaceous reduction of carbon dioxide to the monoxide, which occurs as the temperature in the interior of the burning zone increases as the puff progresses. When the puff ends, the product formation-transmission balance inside the burning zone is interrupted, resulting in a local build-up of gases in their formation regions. These diffuse into the sidestream to deplete the local build-up.

In contrast to the gas phase plume, the sidestream smoke plume originates 0-4 mm behind the paper burn line, becoming visible at temperatures below about 150°C (Fig. 1). This is the approximate position of the tobacco pyrolysis and distillation region inside the cigarette (Baker 1981). Inside the cigarette in this region, a concentrated organic vapour is formed. During the smoulder period much of it will diffuse radially out of the cigarette through the partially degraded cigarette paper, although some will also diffuse axially towards the mouth end of the cigarette to form the smoulder stream. As the vapour diffuses through the paper to the outside, it is subjected to a sudden temperature decrease and dilution. These conditions favour the formation of relatively small aerosol particles compared to mainstream particles.

Since the sidestream plume is the major contributor to ETS, the rate at which sidestream smoke is transported away from the cigarette into the atmo-

a) CO (% v/v)

b) CO (% v/v) AROUND BURNING ZONE, + 3mm FROM PAPER BURN LINE

c) TEMPERATURE (°C)

d) TEMPERATURE (°C) AROUND BURNING ZONE, + 3mm FROM PAPER BURN LINE

e) VERTICAL VELOCITY COMPONENTS IN PLUME 10mm ABOVE CIGARETTE
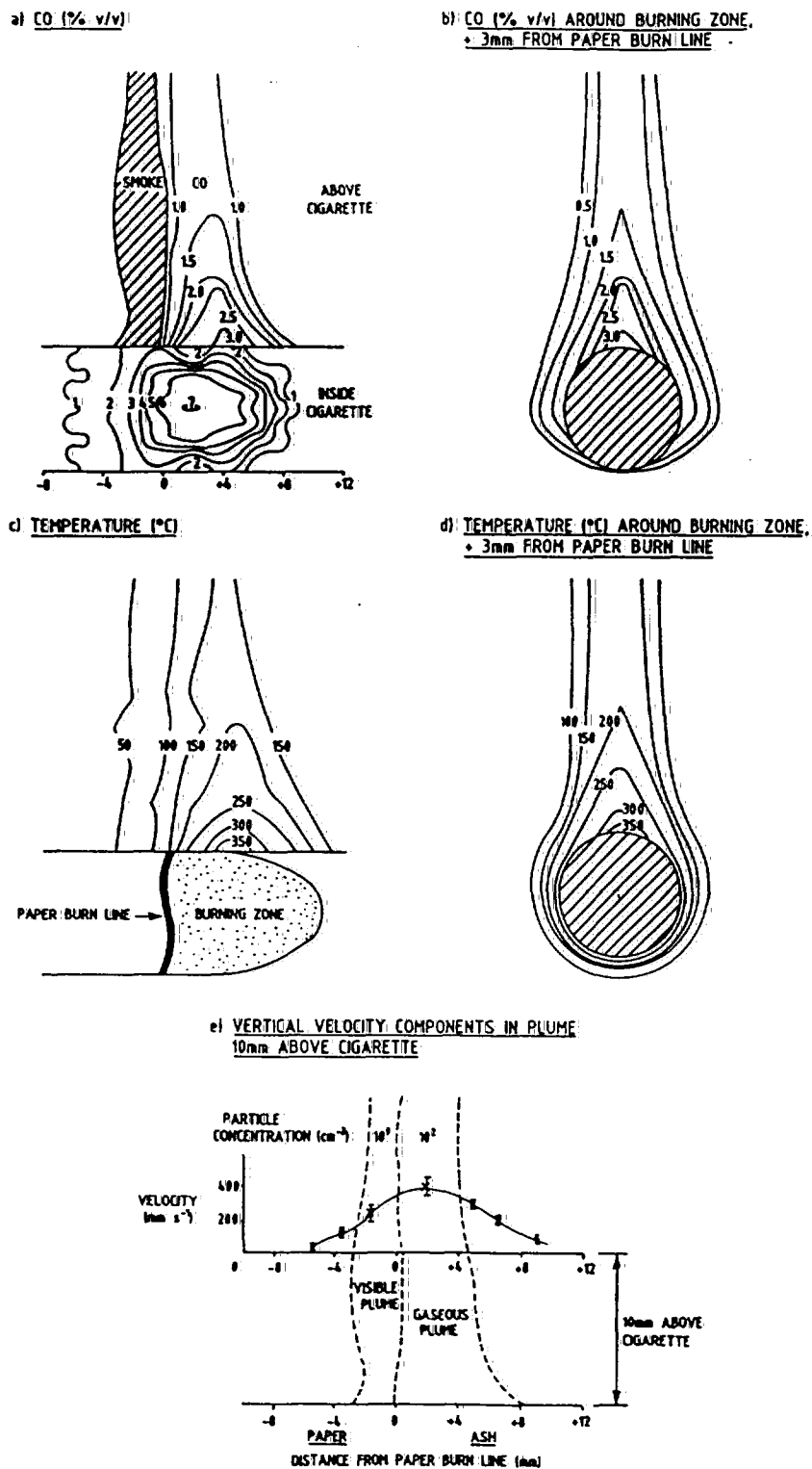
Fig. 1. Concentrations, temperatures and velocities in sidestream plume during smoulder (Baker 1982; Robinson 1987).
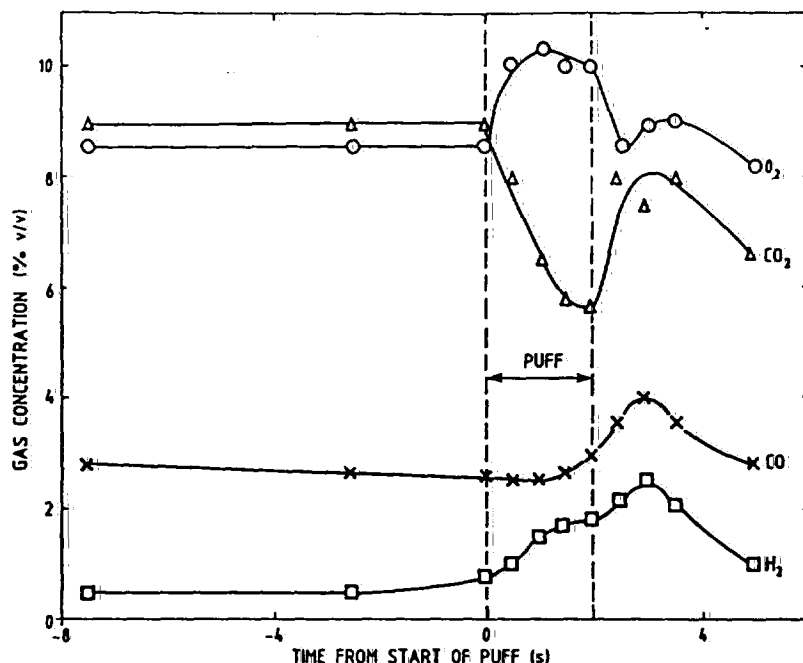
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Fig. 2. Variation with time of sidestream gas concentration 1 mm vertically above cigarette and +3 mm from paper burn line.

sphere ultimately determines the rate of build-up of ETS components. A detailed determination of the velocity distribution of the sidestream plume has been made using a laser Doppler velocimeter described by Robinson (1987). In this technique, two laser beams are focused on a given point in the plume and produce an interference pattern at their point of intersection. Aerosol particles carried in the plume scatter light from the interference pattern and characteristics of this light can be used to calculate the velocity of the particles in the plume. The results indicate that at a given distance above the cigarette there is a distribution of velocity. That obtained at 10 mm above the cigarette is shown in Fig. 1(e) together with an estimate of the particle concentrations in the gas and smoke plumes. The peak velocity of 410 mm s^{-1} occurs in the gaseous plume, 2 mm in front of the paper burn line. The velocities in the smoke plume are generally less than half of this peak velocity.

Measurements at different distances above the cigarette show that the plume is accelerating as it rises above the cigarette (Fig. 3). This acceleration is accompanied by a falling temperature of the plume. The falling temperature of the plume with distance above the cigarette is accompanied by increasing density. This, along with the observed acceleration, means that there is a substantial increase in mass flow rate

in the plume with distance above the plume. This is brought about by air being radially drawn into the buoyancy driven, natural convection sidestream plume as it moves upwards. Detailed calculations and mathematical modelling have confirmed that this occurs (Robinson 1987; Robinson 1988).

Also shown in Fig. 3 are two previous measurements of the velocity of the sidestream smoke plume, that of Neurath et al. (1966) and that of Ayer and Yeager (1982). Both their values were single point measurements, i.e., they did not show any variation of velocity with distance above the cigarette. Clearly the actual flow structure of the sidestream plume is more complex than these two earlier measurements imply.

In general the same chemicals present in mainstream smoke are also present in sidestream smoke, though their relative yield per cigarette is highly dependent on the compound considered. Some typical sidestream/mainstream ratios are shown in Table 1 (Baker 1981; Guerin 1987; Klus and Kuhn 1982; Sakuma et al. 1983, 1984a, 1984b; Norman et al. 1983; Umemura et al. 1986). These ratios were obtained from different studies using different tobaccos, cigarette types, methods of collecting the sidestream smoke, and air movement conditions around the cigarette. These differences will affect the results, reflected by the quoted range for a given compound in
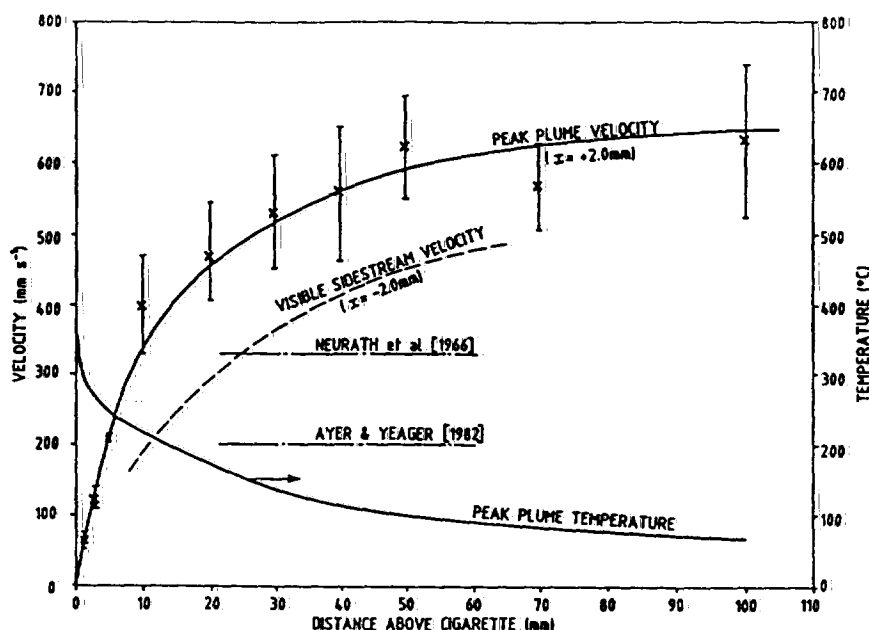
(x is axial distance from paper burn line)

Fig. 3. Measured flow development in the sidestream plume (Robinson 1987).

Table 1. Some typical sidestream/mainstream (SS/MS) yield ratios.

Smoke Component	SS/MS
Hydrogen cyanide	0.06 - 0.5
Succinic acid	0.4 - 0.6
Hydroquinone	0.7 - 1.0
Neophytadiene	1.1 - 1.8
Phenol	1.6 - 3.0
Nicotine	1.9 - 3.3
Acetic acid	1.9 - 3.9
Carbon monoxide	2.5 - 4.7
Benzo(a)pyrene	2.7 - 3.4
Nitric oxide	3 - 13
Limonene	4 - 12
Toluene	5.6
Carbon dioxide	8 - 11
Acrolein	8 - 13
Pyrrole	9 - 14
Naphthalene	17
Pyridine	10 - 20
Water	30
Ammonia	44 - 170
Nitrogen	>270

the table. However, the data do show that there is a very wide range of ratios, varying from 0.06 - 0.5 for hydrogen cyanide to over 270 for molecular nitrogen formed chemically from the tobacco. Of course, one to four times more tobacco is burnt in smoulder than during puffing, depending on the cigarette parameters. Thus, some of the ratios in Table 1 (e.g., phenol and nicotine), simply reflect the proportions of tobacco burnt to the two smoke streams. However, there are many substances which distribute themselves so predominantly to one stream or the other that the reason cannot be due to differences in tobacco consumption. The reasons lie in the different conditions of temperature and mass transfer rates existing in the cigarette burning zone during smoulder and puffing, and the exact mechanism by which different components are formed or released from the tobacco.

During a puff, air is drawn into the peripheral regions of the burning zone, solid phase temperatures in excess of 900°C are reached and it is largely the periphery of the cigarette that burns (Baker 1981). When the puff ceases, the periphery of the burning

zone cools rapidly to about 600°C, air is converted into the back of the burning zone and the axial portion of the tobacco rod is preferentially consumed during about the first twenty seconds of smoulder. The temperatures in the centre of the burning zone are in the region of 800°C and only increase by 50 - 80°C as the puff progresses. The major combustion products, carbon dioxide, carbon monoxide and water, are formed in the high temperature (> 500°C) region of the burning zone. However, the vast majority of smoke species are formed by pyrolysis/distillation processes in a relatively low temperature (< 500°C) oxygen deficient region, just behind the combustion zone in the region of the paper burn line. The concentrated organic vapour so formed is drawn down the tobacco rod during the puff to the mainstream and largely diffuses radially out of the rod during smoulder to form the sidestream smoke.

Hydrogen cyanide is formed via decomposition of nitrates and amino acids. The predominance in the mainstream must reflect a high temperature formation mechanism with sufficient temperatures hardly being attained during smoulder. On the other hand, ammonia, which is formed from the reduction of nitrates and pyrolysis of glycine, is delivered predominantly to the sidestream. Sufficient temperatures must exist during the smoulder period for the pyrolytic generation of the ammonia. Vapour phase water is also delivered almost exclusively to the sidestream and is believed to be derived from oxygen reacting with pyrolytically-generated hydrogen as it diffuses into the sidestream plume (Johnson 1975). Thus, the exact ratio in Table 1 very much depends on the mechanistic origin of each component.

The pH of mainstream and sidestream smoke also differs, with sidestream smoke being generally more alkaline. For example, the pH of the mainstream smoke of a U.S. blended cigarette is typically in the range 6.0 to 6.2 and that of the sidestream is in the range 6.7 to 7.5 (Brunnemann and Hoffmann 1974). In this study, the pH was determined by passing smoke over a sensitive combination electrode connected to a pH meter. The observed sidestream and mainstream difference are due to the predominance of basic components in the sidestream, e.g., ammonia, pyridine and nicotine, and carboxylic acids and phenols in the mainstream smoke. (However, pH is a concept applicable to aqueous solutions and strictly speaking it is not meaningful to give too much significance to the pH of a suspension of aerosol particles).

The mean size of the aerosol particles in sidestream smoke is smaller than in mainstream smoke.

Thus, Okada et al. (1977), using a light scattering technique, reported mainstream particles to have geometric number mean diameter of 0.18 μm and sidestream particles of 0.12 μm. The mean size of the mainstream particles is smaller than that reported in other studies using other measurement techniques, cited by Okada et al. (1977). However, the relative size distributions of the mainstream and sidestream particles are seemingly authentic. The different size distributions for the two smoke streams must reflect the different rates of cooling and levels of air dilution to which their precursor vapours are subjected.

In mainstream smoke, Browne and co-workers (1980), have shown that nicotine is almost entirely in the particulate phase. This also is true for fresh, concentrated sidestream smoke (Proctor 1988a), though nicotine seems to transfer rapidly to the vapour phase as the smoke stream ages and becomes diluted (Eudy et al. 1986; Eatough et al. 1986, 1989).

Sidestream smoke yields will be dependant upon the weight of tobacco burnt during smoking, the construction of the cigarette, and the way in which it is smoked. However, typically, a conventional cigarette will yield around 600 mg of CO_2, 4.5 mg CO, 5 mg of nicotine, and 25 mg particulate matter (water and nicotine subtracted) per cigarette in sidestream smoke.

Exhaled mainstream smoke

Few authors have considered exhaled mainstream smoke (EMS) as anything other than a minor contributor to ETS (Nystrom and Green 1986). Work recently completed in our laboratory suggests that the role of exhaled mainstream smoke should be considered in more detail. A variety of smokers were studied under carefully controlled conditions, smoking three types of cigarettes. These were a typical filtered British flue-cured cigarette (15 mg mainstream particulate delivery), a typical filtered case flavoured U.S. blended cigarette (14 mg), and a low delivery (3 mg) filter ventilated flue-cured cigarette. For each experiment three subjects smoked one cigarette type in their normal manner in a 30 m³ chamber. This was repeated on three occasions for each of the cigarette types. For each experiment the maximum concentrations were determined for particulate matter (as measured by a piezobalance, TSI 5000), nicotine (Tenax trapping, GC-MS analysis) and carbon monoxide (by non-dispersive infrared spectroscopy).

Subsequently, cigarettes were smoked by machine (set at average parameters to mimic the human smoking) with mainstream smoke exhausted from the room. Hence the ETS from sidestream smoke alone was

Table 2. Mean contributions of exhaled mainstream smoke (EMS) to ETS.

Cigarette	Contribution of EMS (%) to		
	ETS Carbon Monoxide	ETS Particulate Phase	ETS Nicotine
Flue-cured	11	43	7
US-blended	13	15	9
Filter Ventilated	3	20	1

compared to ETS produced by humans, including exhaled mainstream smoke.

Table 2 presents the summary data for the three cigarette types. It can be seen that exhaled mainstream smoke contributes little to the gas phase of ETS and that this contribution is dependant upon the mainstream delivery of the cigarettes studied. However, EMS does significantly contribute to the ETS particulate phase. It is thought that particulate matter is retained by the smoker to a greater extent than carbon monoxide, and hence it would be expected that the EMS particulate contribution would be less than observed (Creighton 1973). The results may then indicate that EMS particles are more stable (perhaps larger and containing more water) than fresh sidestream particles.

AGEING OF TOBACCO SMOKE

As sidestream and exhaled mainstream smoke diffuse into the atmosphere and away from the cigarette and smoker, they become ETS. The originally concentrated sidestream and exhaled mainstream smoke streams become greatly diluted, the sidestream smoke cools and accelerates (Figs. 1, 3), and various physical and chemical changes occur in the smoke.

A variety of studies (Eatough et al. 1986, 1987; Eudy et al. 1986; Hammond et al. 1987) have shown that the nicotine in ETS is almost entirely in the vapour phase. Since ETS nicotine originates almost entirely from sidestream smoke (see Table 2), then nicotine in the fresh sidestream particles must rapidly evaporate out of the particles as the smoke ages during initial dilution.

Studies by Pritchard and co-workers (Black et al. 1987; Pritchard et al. 1988) have also shown that matter is evaporated from fresh sidestream particles as they are diluted to form ETS. They loaded 1-iodohexadecane labelled with ^{123}I onto cigarettes. This material has a boiling point of 380°C, typical of that of components found in smoke particles. When fresh sidestream smoke was collected from the smouldering, loaded cigarette using a "fish-tail" chimney collection system described elsewhere (Proctor et al. 1988a) it was found that 5% of the sidestream radioactivity was found in the vapour phase and 95% in the particulate phase. On the other hand when the cigarette smouldered in a steel chamber of 14 m^3 internal volume, 70% of the airborne radioactivity was found to be in the vapour phase, and subsequent radiochemical analysis indicated that there had been no chemical degradation of the 1-iodohexadecane in the environmental chamber. Thus, the material had evaporated out of the sidestream particles during dilution to form ETS.

Ingebrethsen et al. (1985, 1986) have independently estimated that 20 to 30% of the original matter in sidestream particles is lost by evaporation during the ageing of ETS. This estimate was calculated from measurements of the number and sizes of sidestream particles diluted in a stirred 0.5 m^3 stainless steel chamber. The size distribution measurements were made by Ingebrethsen and Sears (1985) using a combination of an optical particle counter, an electrostatic mobility analyser, and a condensation nucleus counter. Sidestream smoke was introduced into the 0.5 m^3 chamber and diluted, and the number and sizes of the particles monitored over six hours. The estimated initial mass loss by evaporation of the smoke particles during about the first hour of ageing resulted in ETS particles with a mean number diameter of 0.098 μm at typical ETS concentrations (several μg m^{-3} particulate matter). This is equivalent to a mass median diameter of 0.185 μm. As the ETS aged over the next few hours in the stirred chamber, there was a slow but gradual increase in mean mass median diameter: 20% increase over 12 hours. This is due to a combination of coagulation of particles and removal of smaller particles by deposition onto surfaces of the chamber. Surface deposition of ETS is the main route of removal in a static environment and

is a function of particle size, mixing rate, room size, and shape.

In addition to physical changes, chemical changes also occur as ETS ages. Thus, for example, nitric oxide slowly oxidises over minutes and hours to nitrogen dioxide in ETS (Piadé and Fink 1987; Klus et al. 1987; Baker et al. 1988). However, these chemical changes, and indeed the particle size changes described above, will be far outweighed by the physical effects of air movement which will occur in real indoor environments. These effects are described in the next section.

BUILD-UP AND DECAY OF ETS CONSTITUENTS

As cigarettes are smoked in a room, the levels of ETS components in the room rise and then fall due to air circulation, room ventilation and, to a lesser extent, interactions of the ETS constituents such as deposition of smoke particles onto surfaces in the room. It is normally not practical to measure the dynamic build-up and decay of ETS constituents in real-life environments and for such information specially constructed environmental rooms are used. In such rooms, the temperature, relative humidity, air circulation rate, and fresh air input can be varied over a wide working range and the observed dynamic ETS levels related to room environmental conditions. A number of studies in such rooms has been published in recent years (Hoegg 1972; Cain and Leaderer 1981; Case 1985; Blake et al. 1986; Heavner et al. 1986; Ingebrethsen et al. 1986; Black et al. 1987; Eatough et al. 1987; Hiller et al. 1987; Olander et al. 1987; Piadé and Fink 1987; Pritchard et al. 1988; Rawbone et al. 1987a, 1987b; Vu Duc and Huynh 1987; Baker et al. 1988) or in standard offices with controlled environments (Klus et al. 1987).

Typical results from one study (Baker et al. 1988) are illustrated in Fig. 4. In this study the ETS was produced from the sidestream smoke of nonventilated filter cigarettes containing flue-cured tobacco smoked under standard smoking machine conditions in a $30 \, m^3$ chamber. The chamber walls and ceiling had an impervious painted plastic finish and the floor was constructed of heavy duty non-slip PVC. Further details of the chamber have been given elsewhere (Case 1985; Baker 1988). Room conditions and number of cigarettes smoked were varied over the following ranges: temperature, 15-30°C; relative humidity, 55-85%; room air circulation, 1-27 air changes per hour; fresh air, 8-25%; number of cigarettes, 2-8.

During an experiment, samples of air were continuously removed from the room and analysed for particulate matter (using a TSI Model 5000 piezobalance fitted with a 3.5 μm filter situated in the room), carbon monoxide and carbon dioxide (using non-dispersive infrared spectroscopy), nitric oxide, and nitrogen dioxide (using chemiluminescence), hydrocarbons (using flame ionisation), and particle size and number distributions (LAS-X laser aerosol spectrometer). For nicotine determination, time weighted average values were obtained over the five minute periods by sampling the air through thermal desorption tubes containing Tenax and subsequent analysis by gas chromatography/mass spectrometry. Room temperature and relative humidity were also monitored during each experiment. The data from all the instruments were monitored on a microcomputer, taking data points every 30 seconds. All results presented are corrected for background readings.

Individual data points are included on some of the profiles in Fig. 4 to indicate the precision of measurement. The signal to noise ratio for most of the instruments was better than 20:1, although that for nitrogen dioxide and total hydrocarbons was 5:1 and 10:1 respectively, due to the analysers being operated at maximum sensitivity. The coefficients of variation of all the profiles over five replicate experiments were better than 5%.

In Figs. 4 and 5, gas concentrations are quoted in $mg \, m^{-3}$ or $\mu g \, m^{-3}$. It is, however, common practice in studies on ETS to use the units 'parts by volume per million' (ppm), which is equivalent to μL/L in the metric system of units, especially for carbon monoxide levels — see, for example, the review by Repace (1987b). The exact conversion factor depends on gas density, and for carbon monoxide at room temperature and pressure, $1 \, mg \, m^{-3} = 1.15 \, ppm$.

All the measured ETS components in Fig. 4 reach a maximum concentration at the end of smoking and then decay exponentially at a first order rate, i.e., decay rate is proportional to component concentration, as indicated by linear $\log_e$ concentration/time plots (not illustrated). The nicotine profile apparently reaches a plateau after it has decayed to about 20% of its maximum value. The profiles for particle number concentration and aerosol mass are almost exactly parallel, as are the profiles for carbon monoxide and carbon dioxide. The count median diameter of the ETS aerosol particles remained constant at 0.13 μm as the ETS built up and decayed over the 5000 seconds of the determination. This is larger than that reported by Ingebrethsen and Sears (1985) in their $0.5 \, m^3$ chamber, due to the laser aerosol spectrometer in the current study not measuring the small particles below 0.1 μm which were included in In-

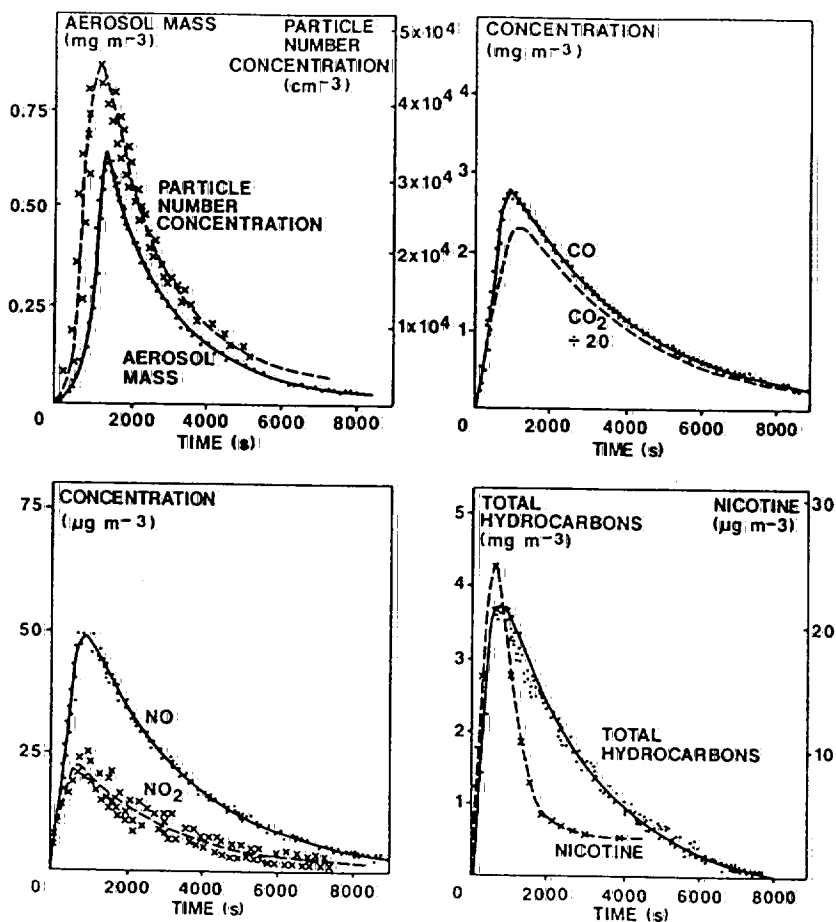9 air changes/hour, 10% ventilation; 20°C, 55% RH, 2 cigarettes smoked

Fig. 4. ETS build-up and decay profiles.

gebrethsen and Sears' combined technique described earlier.

For all the components studied, the peak ETS concentration (or time-weighted average for nicotine) increases linearly with the number of cigarettes smoked for 2 to 8 cigarettes. This linear relationship has been observed previously with up to 30 cigarettes smouldered (Hoegg 1972; Case 1985; Blake et al. 1986). Using two types of experimental cigarettes with filter ventilation levels of 20 and 50%, Blake and co-workers (1986) have demonstrated the linear relationship for the following ETS components: carbon monoxide, nitric oxide, nitrogen dioxide, hydrogen cyanide, ammonia, formaldehyde, phenol, each of the three cresols, nicotine and aerosol particle mass.

The time taken for the peak ETS concentration to decay back to the background level depends on the environmental conditions. Fig. 5 illustrates the effect of air exchange rate (equal to room air circulation rate multiplied by the fraction of fresh air admitted to room) on the decay of carbon monoxide. Clearly, air movement has a large effect on the decay, as expected from mathematical considerations (Repace 1987a; Robinson 1988). The effect of air exchange rates on the half-life times of ETS components, obtained from profiles similar to those in Figs. 4 and 5 is illustrated in Fig. 6 over the sixteen sets of environmental conditions used in the study.

Half-life time is defined as the time to decay to half the value of the maximum concentration. Half-

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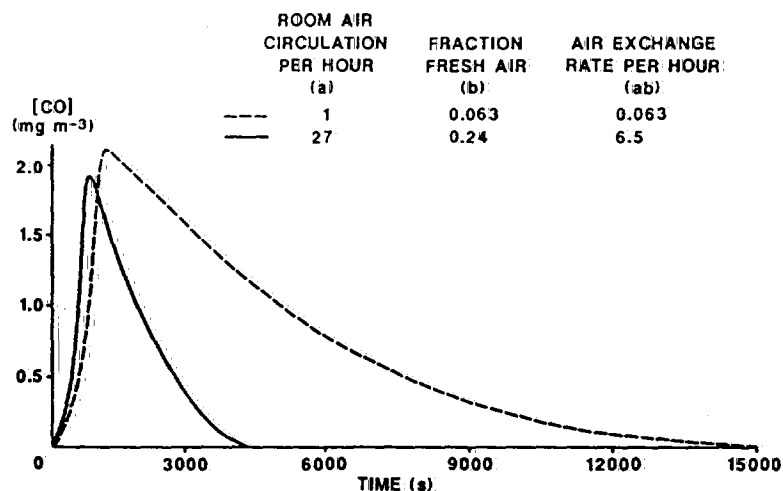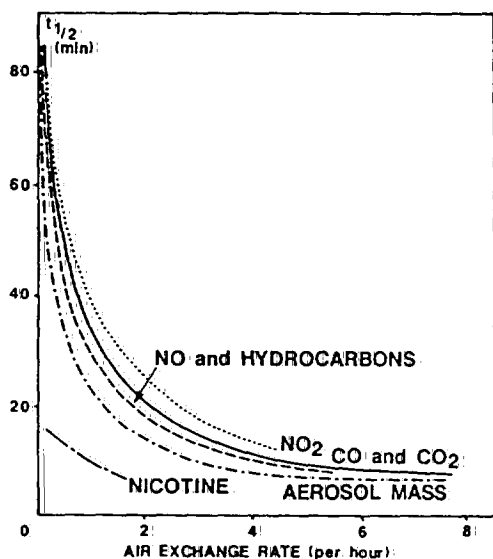|	ROOM AIR CIRCULATION PER HOUR (a)	FRACTION FRESH AIR (b)	AIR EXCHANGE RATE PER HOUR (ab)
----	1	0.063	0.063
——	27	0.24	6.5

Fig. 5. ETS CO profiles for two room conditions.

Fig. 6. Variation of ETS $t_{1/2}$ values with air exchange rate.

life times for nicotine were not obtained under all the conditions in the study, and in general are much smaller than the values for the other components, and much less influenced by the air movement. They are also less accurately defined, since they are based on average levels over five-minute periods rather than continuous measurements, and since nicotine is decaying relatively fast. Similar $t_{1/2}$ values for nicotine can be obtained from the decay profiles reported in other studies: values of 10, 14 and 23 minutes from

Eudy and co-workers' profiles (1986) and 16 and 23 minutes from the profiles of Rawbone and co-workers (1987a). Variation of the environmental temperature (15-30°C) and relative humidity (55-85%) had a negligible effect on the $t_{1/2}$ values (detailed results not illustrated).

For a given set of environmental conditions, the trends in Fig. 6 indicate that the relative $t_{1/2}$ values of the ETS components are in the order:

$NO_2 > CO = CO_2 > NO \approx$ total hydrocarbons > aerosol mass > nicotine
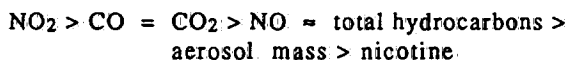
The decay rates of the carbon oxides depend solely on the effects of air change rate and fresh air input (Piadé and Fink 1987) and their levels in ETS remain constant when there is no air movement (Heavner et al. 1986; Hiller et al. 1987). Where a component has smaller $t_{1/2}$ values than carbon monoxide there must be some mechanism other than just air movement depleting its levels — chemical or physical. Nitric oxide $tv_{1/2}$ values are on average significantly less than those for carbon monoxide, and nitrogen dioxide $t_{1/2}$ values are significantly higher, at the 90% confidence level. This is due to the conversion of nitric oxide to nitrogen dioxide in the ambient air, as postulated by others (e.g., Piadé and Fink 1987; Klus et al. 1987).

$$2NO + O_2 \rightarrow 2NO_2 \qquad [1]$$
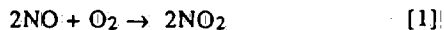
In fact, using the experimental data on ETS nitric oxide and nitrogen dioxide decay levels it can be shown that the rate constant for reaction [1] in ETS

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is almost an order of magnitude higher than that for the pure gas phase conversion but similar to that in the gas phase of mainstream smoke (Baker et al. 1988). Thus, in ETS, and the gas phase of mainstream smoke, the oxidation of nitric oxide is catalysed by substances which are not known.

The smaller values of $t_{1/2}$ for aerosol mass than carbon monoxide must be due to deposition of the particles onto the surfaces in the room. The very much smaller values for nicotine could be due to adsorption of nicotine vapour onto the surfaces — since, as indicated in the preceding section, nicotine in ETS is largely in the vapour phase.

REAL-LIFE LEVELS OF ETS COMPONENTS

Although ETS originates from sidestream and exhaled mainstream smoke, the great dilution and other changes which these smoke streams undergo as they form ETS make their properties significantly different from those of ETS. Thus, the sidestream/mainstream ratios quoted in Table 1 can be misleading if used out of context. The important question is not the ratio of sidestream/mainstream but rather what is the concentration of the constituent in the indoor environment and how does it compare to levels from sources other than ETS. Studies based solely on observations of fresh sidestream, or highly and unrealistically concentrated ETS, should take into account the possible differences between these smokes and ETS found in real-life situations.

The previous sections have described how the concentration of ETS in a room will depend upon many factors such as the number of cigarettes smoked, how they were smoked and what type they were, and on the size and ventilation conditions in the room. The situation is further complicated in the real world by the fact that ETS is only one contributor to indoor air containing chemicals arising from multiple sources (Proctor et al. 1988b). All indoor air environments contain numerous chemicals as a result of emissions from, for example, building materials, furnishings, cooking and heating fuels, and consumer products (NRC 1981). Many of the chemicals associated with ETS will also be present as a result of such sources (Jenkins and Guerin 1984; Proctor et al. 1988a, 1989a, 1989b).

Hence, in order to determine potential exposures to ETS, it is essential to employ methods that allow a distinction between substances present as a result of tobacco smoking and substances present as a result of the other various sources. One approach is to identify a chemical marker that is specific, or at least indicative, to ETS (Haley et al. 1988).

Much early research used carbon monoxide concentrations to assess ETS levels (Sterling and Dimas 1982). However, there are many sources of carbon monoxide, such as gas cookers or heaters, open fires, or motor vehicle emissions drawn in from outdoors, and it is not possible in real-life situations to segregate the ETS contribution from this background (NRC 1981; Girman and Traynor 1983; Haley et al. 1988).

Nicotine is far more specific, and had been used in recent years. The presence of nicotine in air is almost certainly indicative that tobacco smoking is, or has been taking place. Typically indoor levels range from 5 to 70 $\mu g\ m^{-3}$.

However, it should be noted that the behaviour of nicotine in ETS is somewhat unusual when compared to many of the other constituents. Nicotine is primarily a vapour phase constituent in ETS, though a small portion (around 2%) will be found associated with the particulate phase (Eatough et al. 1989). Research has shown that the nicotine decays rapidly from an atmosphere in comparison even to other vapour phase constituents of ETS (Nystrom and Green 1986; Eatough et al. 1989 and Fig. 4 of the present paper). Moreover, it is also likely that some nicotine adsorbed onto walls and furnishings will be re-emitted. If this were to be the case, then areas where smoking had not taken place for some time might still exhibit a low level of airborne nicotine.

Even so, nicotine is currently the most useful specific chemical marker for ETS, and many field studies have utilised its measurement. For example, Thompson et al. (1989) found airborne nicotine concentrations in restaurants ranged from 0.5 to 37.2 $\mu g\ m^{-3}$ with a geometric mean of 3.5 $\mu g\ m^{-3}$. In offices, Hammond et al. (1987) found personal exposures to nicotine (nonsmokers) ranging from 3.1 to 28.2 $\mu g\ m^{-3}$, whilst Carson and Erikson (1988) using fixed site monitoring, in a study of 31 offices in Ottawa found airborne nicotine geometric mean to be 7.2 $\mu g\ m^{-3}$ (range <1.2 to 69.7 $\mu g\ m^{-3}$). Research undertaken by our laboratory found similar levels of airborne nicotine with a median of 3.1 $\mu g\ m^{-3}$ (range 0.6 to 26 $\mu g\ m^{-3}$) in smokers' offices, 15.5 $\mu g\ m^{-3}$ (range 0.6 to 49.3 $\mu g\ m^{-3}$) in smoking-allowed train compartments, and 18 $\mu g\ m^{-3}$ (range 3 to 57 $\mu g\ m^{-3}$) in betting shops (Proctor et al. 1989a, 1989b).

Respirable suspended particulates (RSP) have also been determined in indoor air in relation to ETS. Virtually all ETS particulate matter will be in the respirable fraction (i.e., less than 3.5 μm diameter) of airborne particulate matter.

However, in real-world situations, ETS will seldom be the sole source of particulate matter. Hence it is important to try and estimate the proportion of particulate matter relating directly to ETS, rather than just measuring total particulate matter. One of the most frequently referenced papers is the work of Repace and Lowrey (1985). Their research suggested that a typical nonsmoker working in an office building in the U.S. would be exposed daily to average concentrations of particulate matter due specifically to ETS of 242 μg m^{-3} (range 100 to 1000 μg m^{-3}). However, Samet et al. (1987) stated that surveys of indoor air quality based on measurement of total suspended particulate concentrations (such, presumably, as the Repace and Lowrey study) will not readily identify the excess mass indoors from environmental tobacco smoke.

Some researchers have attempted to develop more specific methodologies for the determination of the ETS proportion of total RSP. One method extracts respirable particulate matter collected on teflon coated filter pads, and subsequently analyses the extracts for their ultra-violet (UV) absorbance at 325 nm. By using a surrogate standard, $2,2^1,4,4^1$-tetrahydroxybenzophenone, calibrated against ETS formed in controlled conditions, an estimate of the ETS contribution to particulate matter can be made (Thomas et al. 1989). This measure has been termed UV-RSP, and will often be an over-estimate, as smoke of the particulate matter in indoor air originating from sources other than ETS will also result in UV absorbance.

An alternative method, based on the same philosophy, analyses the methanol extracts of collected particulate matter for fluorescence (Thomas et al. 1989). Sample solutions are calibrated against dilute mainstream tobacco smoke solutions, and the method is suggested to have a greater sensitivity and selectivity than the UV-RSP measure.

A more specific particulate marker may be the compound solanesol. This substance is present in relatively high levels in tobacco and is transferred intact to sidestream smoke (Ogden and Maiolo 1988). However, it should be noted that different types of tobacco contain different levels of solanesol and hence the ambient concentration will be dependant not only on the number of cigarettes smoked but also, to some extent, on which brand of cigarettes was smoked.

Relatively few studies have published data using these particulate partitioning methods. Our data from offices, train compartments, and betting shops (Proctor et al. 1989a, 1989b) suggest that, although ETS does add to the particulate levels in indoor environments, it may not always be the predominant source, and is unlikely to be the sole source. In smokers' offices a median RSP level of 91 μg m^{-3} (range 33 to 260 μg m^{-3}) was found, though the corresponding UV-RSP data (the estimate of ETS contribution) gave a median value of 24 μg m^{-3} (range 0.5 to 75 μg m^{-3}). This would suggest that in a relatively well ventilated office environment, ETS was contributing approximately, on average, 26% to the total RSP level. In smoking allowed-train compartments, a median RSP level of 249 μg m^{-3} (range 71 to 325 μg m^{-3}) was identified. The corresponding UV-RSP data here were a median of 70 μg m^{-3} (range 13 to 110 μg m^{-3}). This would suggest an approximate 28% contribution of ETS to total particulate matter. In betting shops (smoking allowed) the median RSP was 284 μg m^{-3} (range 73 to 767 μg m^{-3}) whilst the median UV-RSP was 109 μg m^{-3} (range 57 to 610 μg m^{-3}); an approximate ETS contribution of 38%. This research suggests the necessity for future studies to attempt to apportion ETS particulate matter from total particulate matter.

Although ETS contains many chemicals, research has been unable to identify good chemical markers apart from nicotine and ETS-specific RSP measures. Eatough et al. (1989) have suggested that 3-ethenylpyridine, myosamine, nitrous acid and pyridine may be possible markers for ETS gas phase, though more research is necessary to determine the validity of these substances.

An alternative approach to determining the contribution of ETS to an environment is to compare directly smoking and nonsmoking situations. In order to do this successfully, it is essential to match closely the factors impacting on the environments; for example smoking and nonsmoking situations should be of similar size, ventilation conditions, occupancy, furnishings, etc. This may be achieved by selecting sites within the same building, or by taking large numbers of sites for a particular environmental category. For example, Spengler et al. (1981) measured total RSP values in 80 homes to conclude that a smoker of 20 cigarettes per day would contribute 20 μg m^{-3} to 24-hour indoor particulate concentrations. This type of approach is most usefully undertaken using ETS specific markers, but it may also identify differences in the levels of nonspecific chemicals associated with ETS. In the U.S. Environmental Protection Agency's Total Exposure Assessment Methodology (TEAM) study, levels of some airborne volatile organic chemicals (VOC) were suggested to be higher in smokers' homes than in nonsmokers' homes (Wallace et al. 1987). Other small studies have been unable to dis-

TOTAL ION COUNT
100%

ONE SMOKER IN
OFFICE

NICOTINE

TOTAL ION COUNT
100%

NO SMOKERS IN
OFFICE

NICOTINE

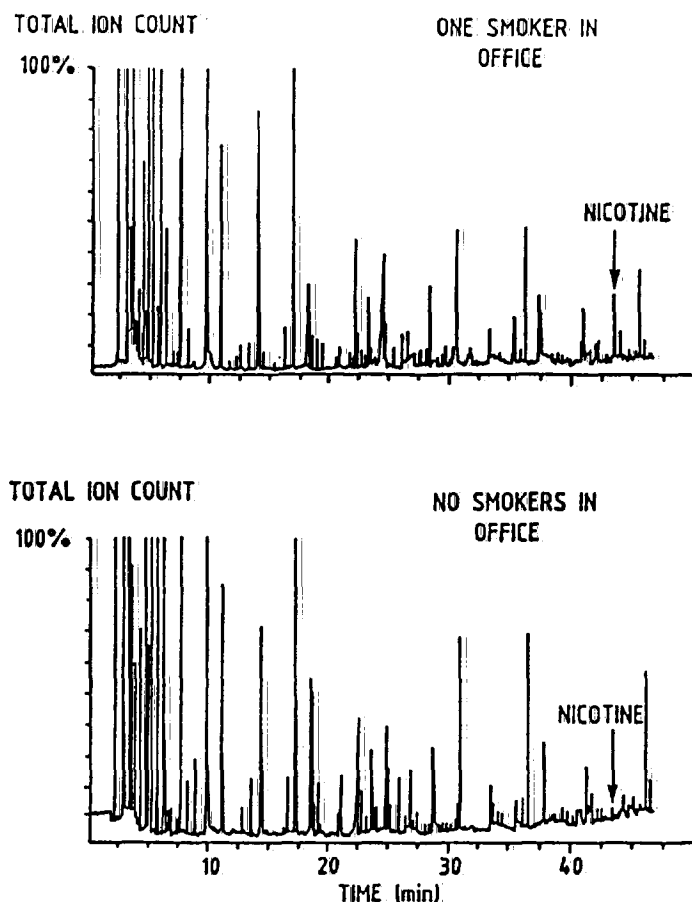10   20   30   40
TIME (min)

Fig. 7. Chromatographic profiles of the air in a smokers' and a nonsmokers' office in a modern, air-conditioned building.

tinguish between the VOC levels in smokers' and nonsmokers' offices (Bayer and Black 1987; Proctor et al. 1989b).

Fig. 7 illustrates a comparison of volatile chemicals in the air of a smoker's and a nonsmokers' office in the same building. The chromatographic profiles were acquired by drawing air through a sampling tube containing Tenax TA adsorbent for an hour sampling period. VOCs are trapped on the Tenax, and recovered for analysis by subsequent thermal desorption — capillary gas chromatography — mass spectrometry (Proctor et al. 1988b). The two offices were virtually identical, apart from the presence of one smoker (who smoked 3 cigarettes during the sampling period), and three nonsmokers in what is termed the smoker's office, and the presence of four nonsmokers in the other office. The nicotine peak in the smoker's office corresponded to a level of $6\ \mu g\ m^{-3}$. Apart from nicotine, a detailed analysis, using mass

spectrometry revealed a similar range of volatile organic compounds (such as benzene and styrene) and similar concentrations of these chemicals in both the smoker's and nonsmokers' office. This finding is not surprising, as the nicotine peak would have to dominate the chromatographic profile in order that the VOC contribution from ETS could be detected above the background level (nicotine being the predominant ETS-associated volatile). A small amount of nicotine ($0.5\ \mu g\ m^{-3}$) was found in the nonsmokers' office, but the similarity in VOC levels between smoking and nonsmoking situations is not due to recirculation of chemicals from smoker's offices because both UV-RSP and nicotine levels in the nonsmoking areas were far lower (by a factor of at least 10) than in smoking areas.

Hence, the accurate assessment of ETS in real-life situations relies upon the use of specific chemical markers to distinguish the ETS contribution from the

chemical background, arising from various sources, that is present whether smoking is taking place or not.

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